

Scattering and Diffraction of Electromagnetic Radiation: An effective probe to material structure

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Scattered electromagnetic waves from material bodies of different forms contain, in an intricate way, precise information on the intrinsic, geometrical and physical properties of the objects. Scattering theories, ever deepening, aim to provide dependable interpretation and prediction to the complicated interaction of electromagnetic radiation with matter. There are well-established multiple-scattering formulations based on classical electromagnetic theories. An example is the Generalized Multi-particle Mie-solution (GMM), which has recently been extended to a special version – the GMM-PA approach, applicable to finite periodic arrays consisting of a huge number (e.g., $\gg 10^6$) of identical scattering centers [1]. The framework of the GMM-PA is nearly complete. When the size of the constituent unit scatterers becomes considerably small in comparison with incident wavelength, an appropriate array of such small element volumes may well be a satisfactory representation of a material entity having an arbitrary structure.

X-ray diffraction is a powerful characterization tool used in a variety of scientific and technical fields, including material science. A diffraction pattern is nothing more than the spatial distribution of scattered intensity, determined by the distribution of scattering matter by way of its Fourier transform [1]. Since all linear dimensions entered into Maxwell's equations are normalized by wavelength, an analogy exists between optical and X-ray diffraction patterns. A large set of optical diffraction patterns experimentally obtained can be found in the literature [e.g., 2,3]. Theoretical results from the GMM-PA have been scrutinized using a large collection of publically accessible, experimentally obtained Fraunhofer diffraction patterns. As far as characteristic structures of the patterns are concerned, theoretical and experimental results are in uniform agreement; no exception has been found so far.

Closely connected with the spatial distribution of scattered intensities are cross sections, such as for extinction, scattering, absorption, and radiation pressure, as a critical type of key quantity addressed in most theoretical and experimental studies of radiative scattering. Cross sections predicted from different scattering theories are supposed to be in general agreement. For objects of irregular shape, the GMM-PA solutions can be compared with the highly flexible Discrete Dipole Approximation (DDA) [4,5] when dividing a target to no more than $\sim 10^6$ unit cells. Also, there are different ways to calculate the cross sections in the GMM-PA, providing an additional means to examine the accuracy of the numerical solutions and to unveil potential issues concerning the theoretical formulations and numerical aspects.

To solve multiple scattering by an assembly of material volumes through classical theories such as the GMM-PA, the radiative properties of the component scatterers, the complex refractive index in particular, must be provided as input parameters. When using a PA to characterize a material body, this involves the use of an adequate theoretical tool, an effective medium theory, to connect Maxwell's phenomenological theory with the atomistic theory of matter. In the atomic theory, one regards matter as composed of interacting particles (atoms and molecules) embedded in the vacuum [6]. However, the radiative properties of atomic-scaled particles are known to be substantially different from bulk materials. Intensive research efforts in the fields of cluster science and nanoscience attempt to bridge the gap between bulk and atom and to understand the transition from classical to quantum physics. The GMM-PA calculations, which place virtually no restriction on the component-particle size, might help to gain certain insight into the transition.

1. Y.-L. Xu, *J. Opt. Soc. Am. A* **30**, 1053 (2013); **31**, 322 (2014); **32**, 12 (2015).
2. G. Harburn, C.A. Taylor, T.R. Welberry, *Atlas of Optical Transforms* (Cornell University Press, 1975).
3. A. Lipson, S.G. Lipson, and H. Lipson, *Optical Physics* (Cambridge University Press, 2011).
4. E.M. Purcell and C.R. Pennypacker, *Astrophys. J.* **186**, 705 (1973).
5. B.T. Draine, *Astrophys. J.* **333**, 848 (1988); B.T. Draine and P. Flatau, *J. Opt. Soc. Am. A* **11**, 1491 (1994).
6. M. Born and E. Wolf, *Principles of Optics*, 7th ed. (Cambridge University Press, 1999).



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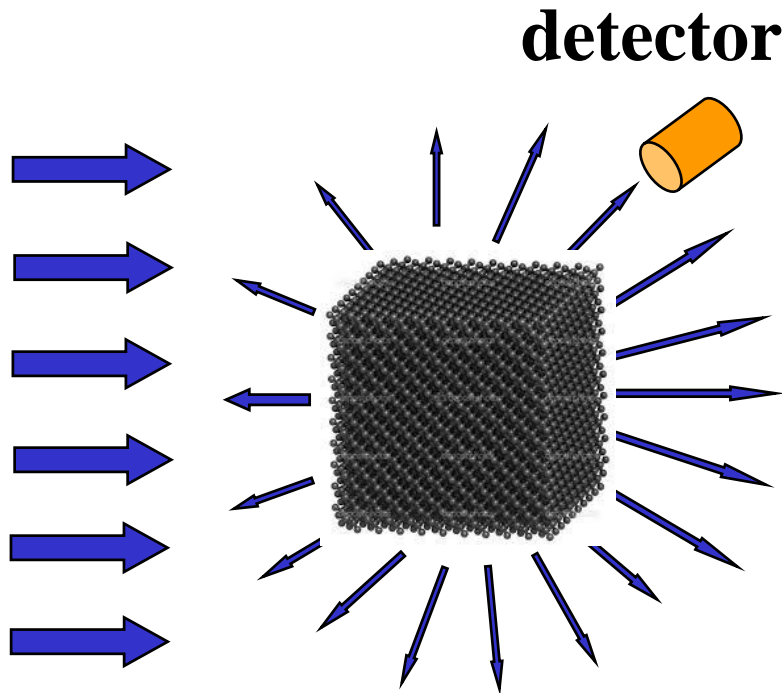
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23-27 October 2016



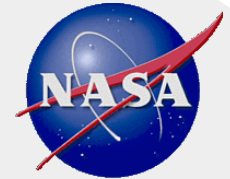
The Intricate Interaction of Electromagnetic Radiation with Materials: “Light Scattering”



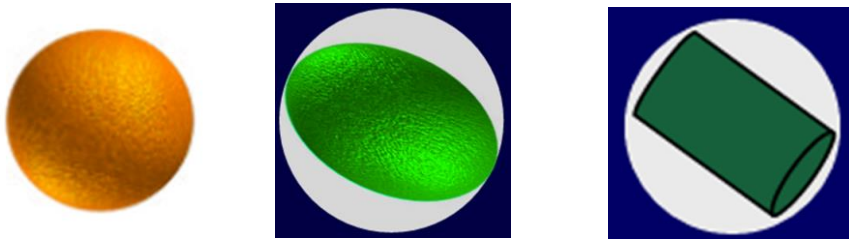
- Incident-wavelength dependent
- Coherent and incoherent light
- Laser, X-ray, and crystal scattering
- Elastic (classical) and quasi-elastic (dynamical) or inelastic scattering

Considering the molecular nature of matter, everything except vacuum is heterogeneous in some sense. In solids and liquids, molecular separation is $\sim 2\text{-}3 \text{ \AA}$, whereas for gases at standard temperature and pressure it is $\sim 30 \text{ \AA}$.

- **Scattered (and diffracted) radiation carries information on the nature of the scatterers**
 - Size, shape, structure, spatial orientation ...
 - Material composition, basic structure ...
 - The information is embedded in the spatial distributions of intensity, polarization, and phase of the scattered radiation
- **Extinction, absorption, radiation pressure ...**



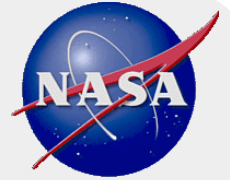
Single-Body Scattering: Rigorous Solutions



- Mie Theory (Lorenz 1898, Mie 1908)
- T -matrix solution (Waterman 1971)
- Other theories

Any rigorous scattering theory for objects with irregular shape and structure ?



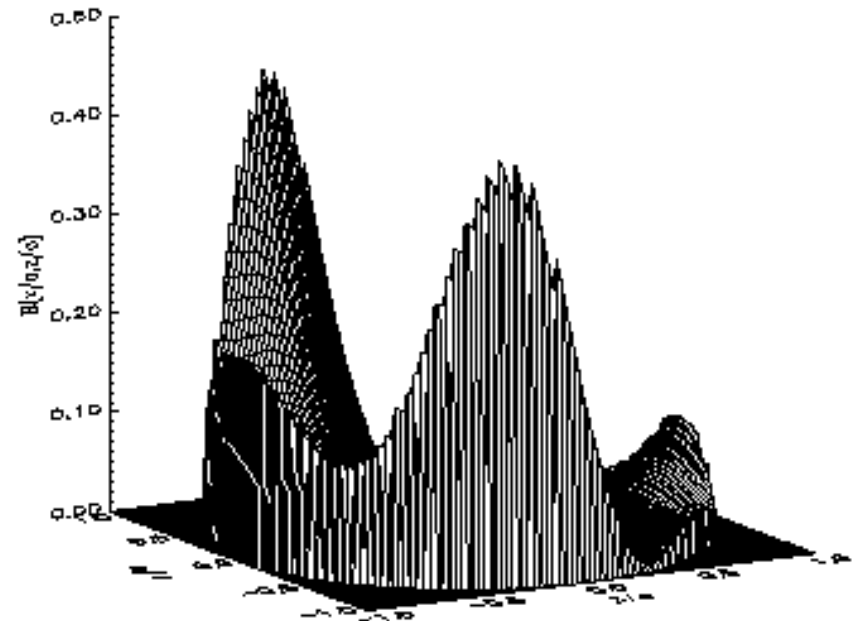
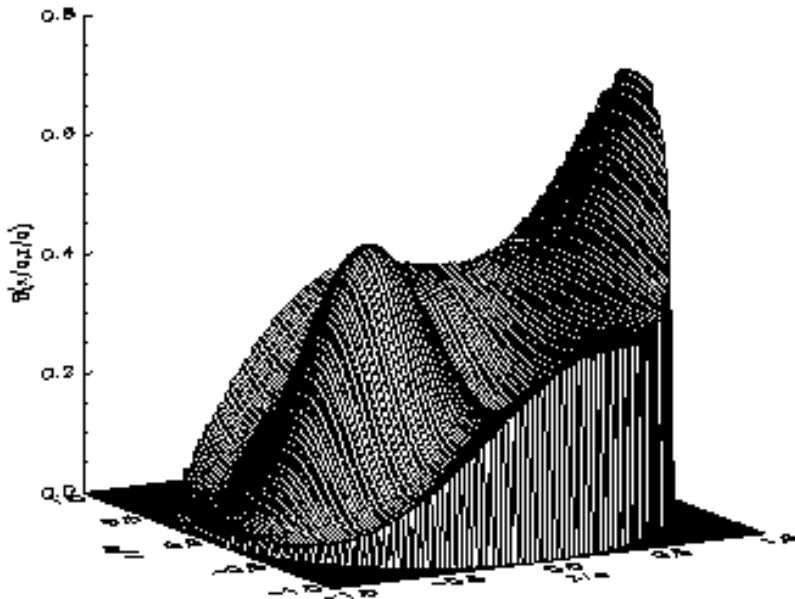


Mie-Predicted Internal Field Distributions

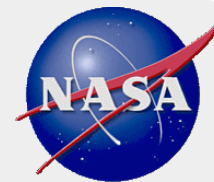
refractive index: (1.95, 0.66)

sphere size-parameter: 2.0

sphere size-parameter: 5.0

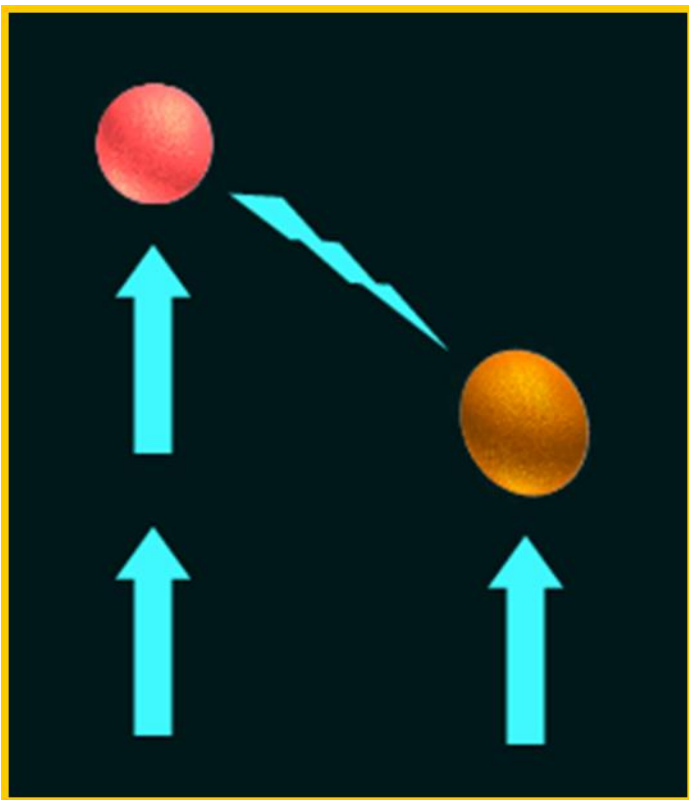


- Heat source functions; Light-dependent Brownian motion of small particles
- Absorption: part of incident electromagnetic energy may be transformed into other forms (thermal, for example)

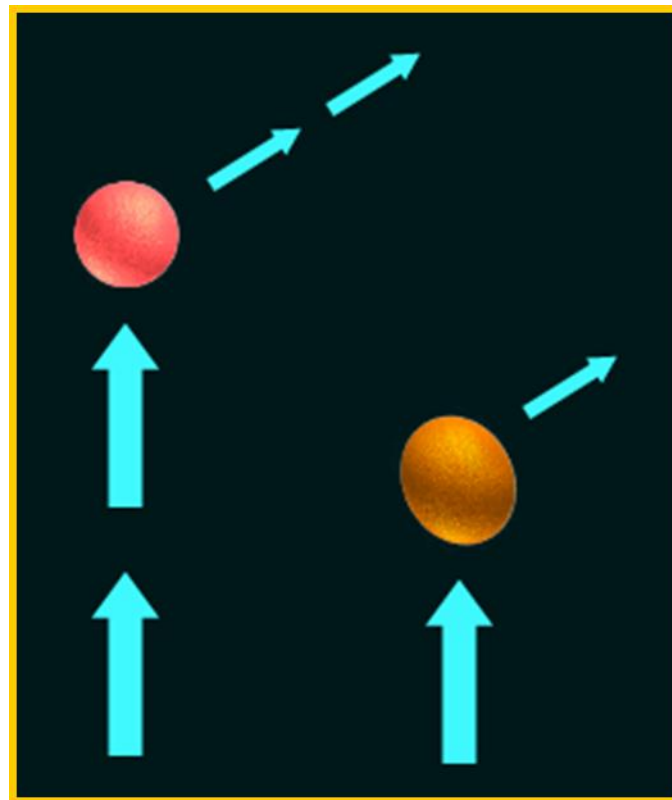


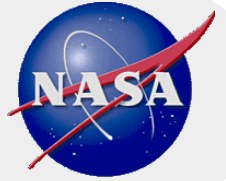
Multiple Scattering Effects: Dependent and Independent Scattering

Interaction



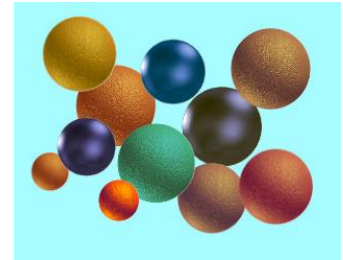
Interference





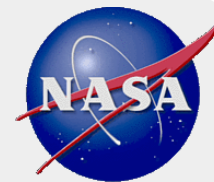
Public-Domain Theoretical Tools and Computer Codes for Multi-body EM-Scattering Calculations

- ✓ Mie-type multi-particle scattering theories
 - **Multiple-Sphere T -matrix Method (MSTM)**
 - For sphere clusters
 - **Generalized Multi-particle Mie-solution (GMM)**
 - For an arbitrary ensemble of scattering bodies that can have arbitrarily mixed size, shape, material composition, and structure
- ✓ Finite-difference time-domain (FDTD) ...
- ✓ (DDA: discrete-dipole approximation)
 - Works better for dielectric objects

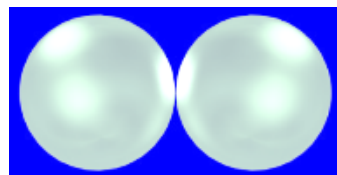


arbitrary
overall shape
and structure
in 2-D or 3-D

Online available MSTM and GMM codes:
(MSTM) <http://www.eng.auburn.edu/~dmckwski/scatcods>
(GMM) <http://code.google.com/p/scatterlib>



Comparison of GMM with Microwave Analog Scattering Measurements: Example 1

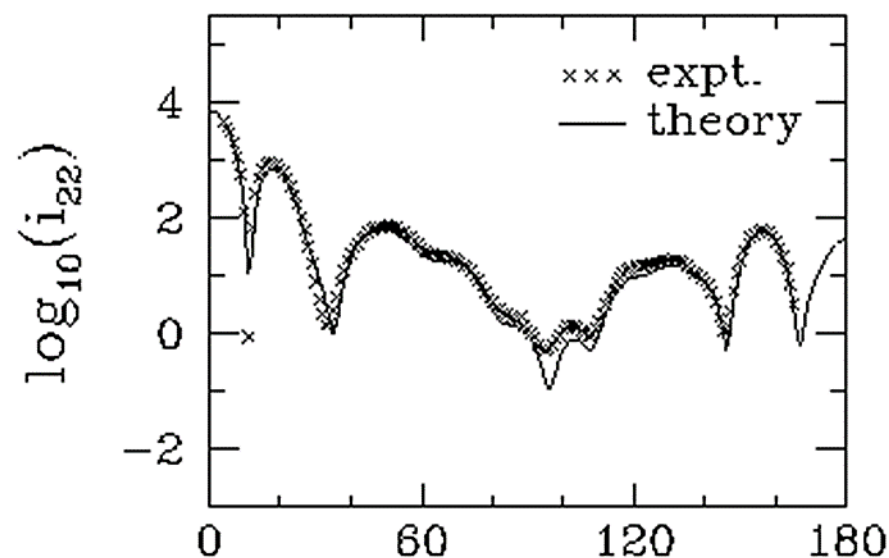
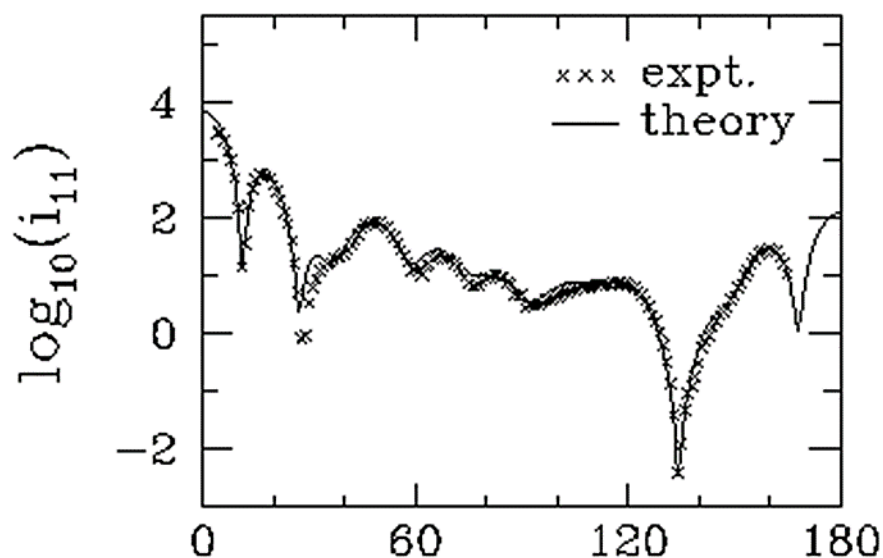


two identical, touching, BK7 optical glass spheres

incident wavelength: 4 mm

single-sphere size-parameter: 7.86

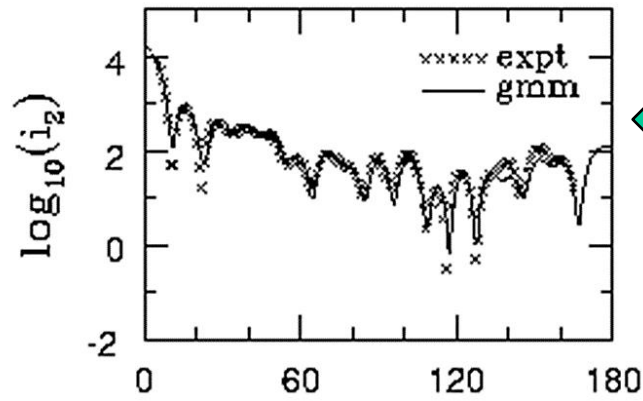
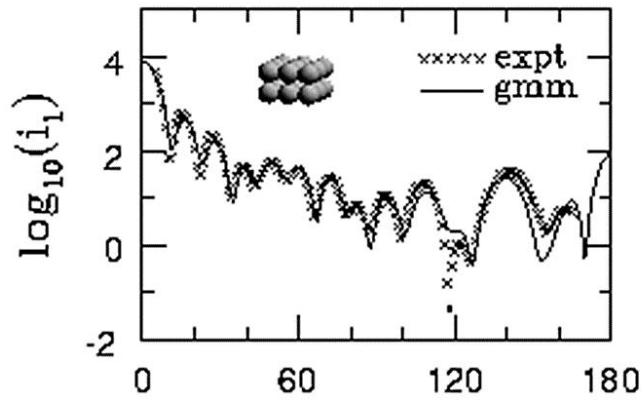
refractive index of sphere: (2.5155, 0.0213)



Scattering Angle (degrees)



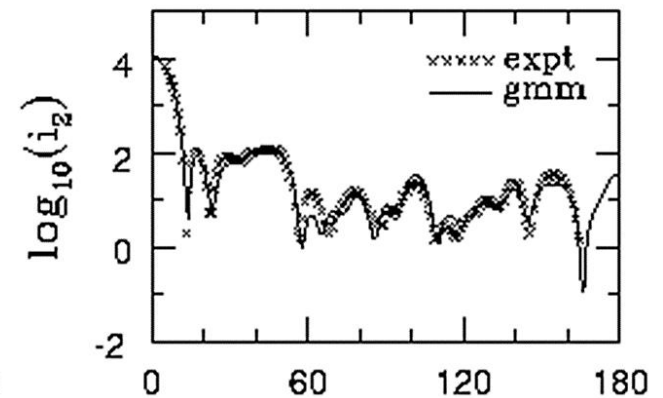
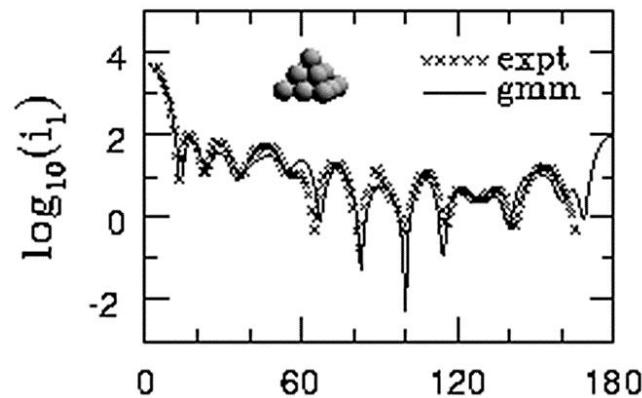
Comparison of GMM with Microwave Analog Scattering Measurements: Example 2



← a rectangular
array of 18
identical spheres

Scattering angle (degrees)

an array of
14 identical
spheres →

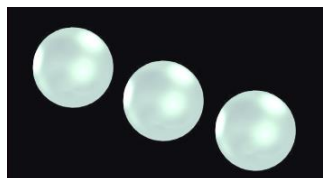


Scattering angle (degrees)



Comparison of GMM with Microwave Analog Scattering Measurements: Example 3

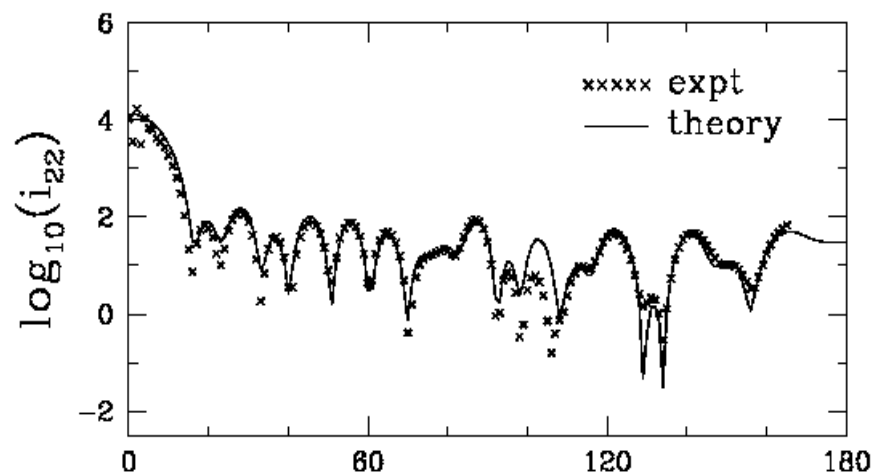
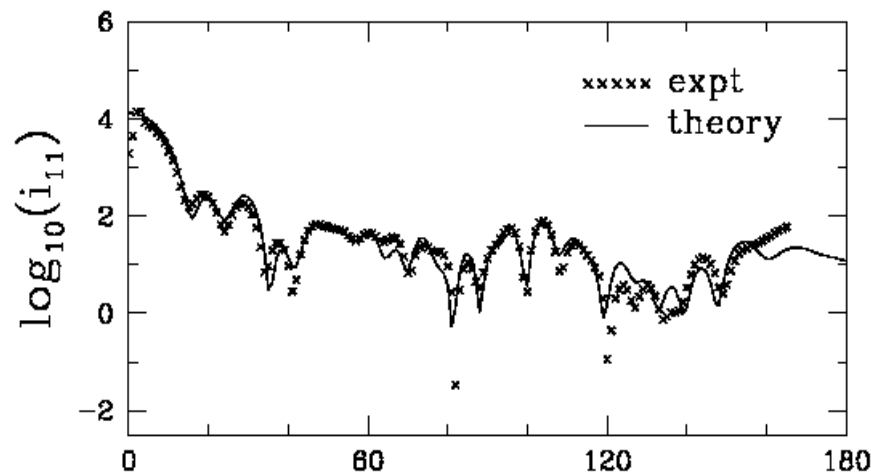
a 15-sphere
array



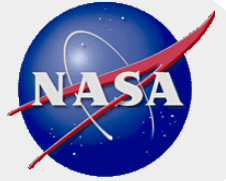
size parameter of a
sphere: 7.49
refractive index:
(2.516, 0.0213)



size parameter of a
sphere: 5.03
refractive index:
(1.615, 0.008)



Scattering Angle (degrees)

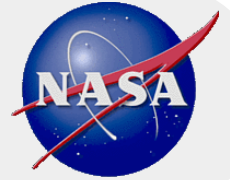


Inevitable Limitations of GMM and MSTM in Practical Implementations

The total number of scattering components allowed in an aggregation: $N \ll \sim 10^5$

COMPUTING TIME $\sim N^2$, where N is the total number of scattering units in an array

An array with $\sim 10^4$ wavelength-sized particles (e.g., whose individual size parameter is ~ 1) requires a couple of days to complete the scattering calculations for a single, fixed spatial orientation using the DELL desktop computer I use. When the total number of the component particles increases to $\sim 10^5$, just 10 times larger, it would demand more than six months to complete the same type of calculation.

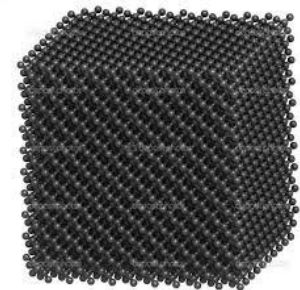
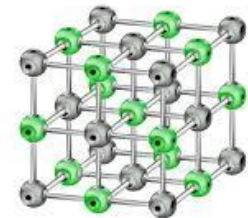
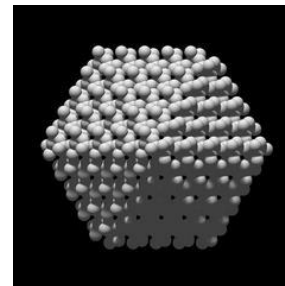
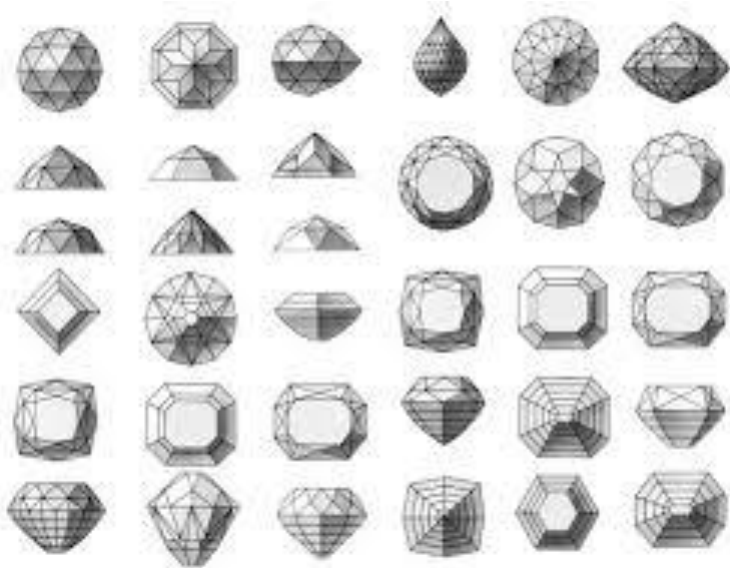
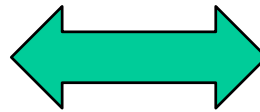


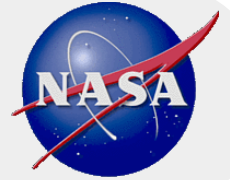
GMM-PA: A Special Version of GMM

- **A special version of GMM specifically for periodic arrays (PAs) of identical scattering units has been developed recently**
 - ✓ Y.-L. Xu, “Scattering of electromagnetic waves by periodic particle arrays,” J. Opt. Soc. Am. A **30**, 1053-68 (2013).
 - ✓ Y.-L. Xu, “Scattering of electromagnetic radiation by three-dimensional periodic arrays of identical particles,” J. Opt. Soc. Am. A **31**, 322-331 (2014).
- **The main purpose of deriving the GMM-PA formulations is to make it possible to calculate aggregations with a huge number of components that standard multiple scattering theories are unable to handle**
- **The PA-approach is highly efficient, as regards computing power-resource and time requirements**
 - Containing an approximation with respect to “edge effect,” negligible for aggregates with a sufficiently large number (e.g., $\gg 10^6$) of components



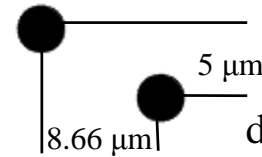
GMM-PA





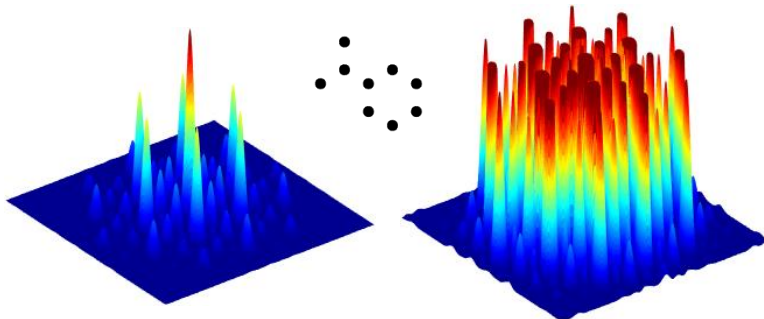
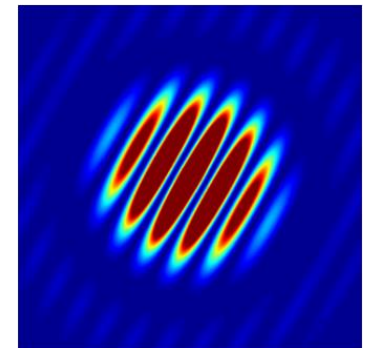
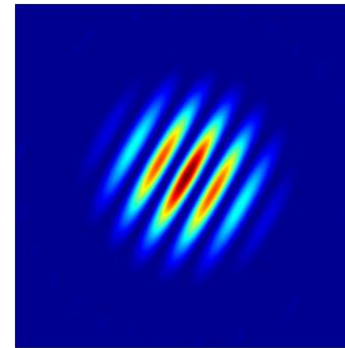
Fraunhofer Diffraction Patterns

As a part of experimental test, GMM-PA predicted Fraunhofer diffraction patterns are systematically compared with experimental results published in “**ATLAS OF OPTICAL TRANSFORMS**” (G. Harburn, C.A. Taylor, and T.R. Welberry)

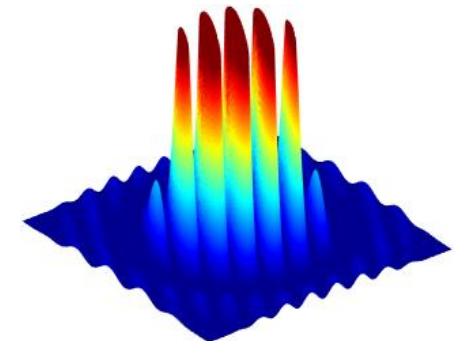
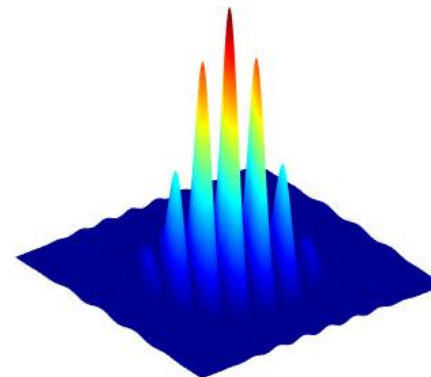


wavelength = 633 nm
(red He-Ne Laser)

diameter = 3 μm



(Plate 4, Pattern 3)



(Plate 2, Pattern 3)

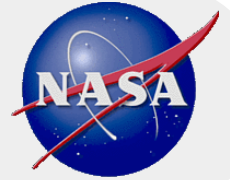
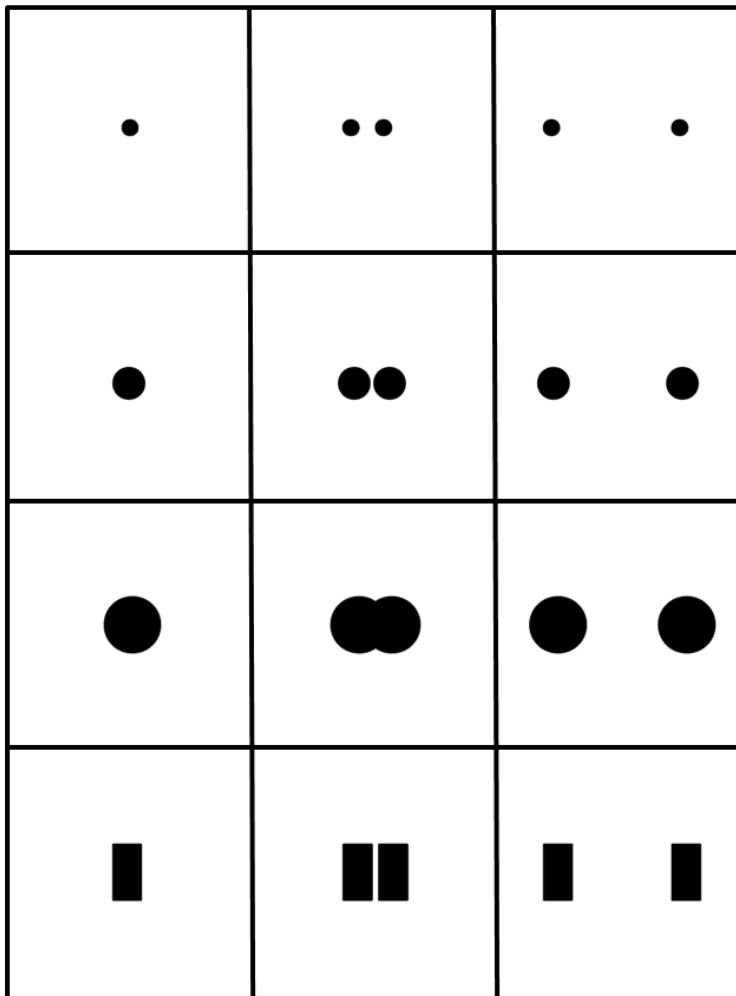
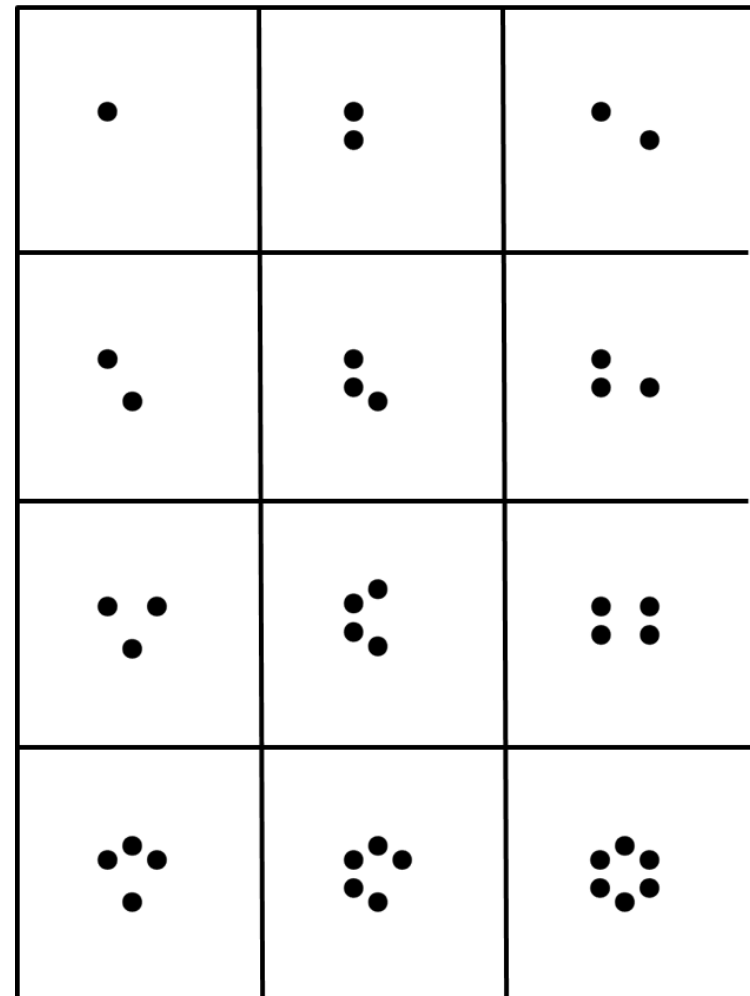


Plate 1 & 2

(a) Plate 1

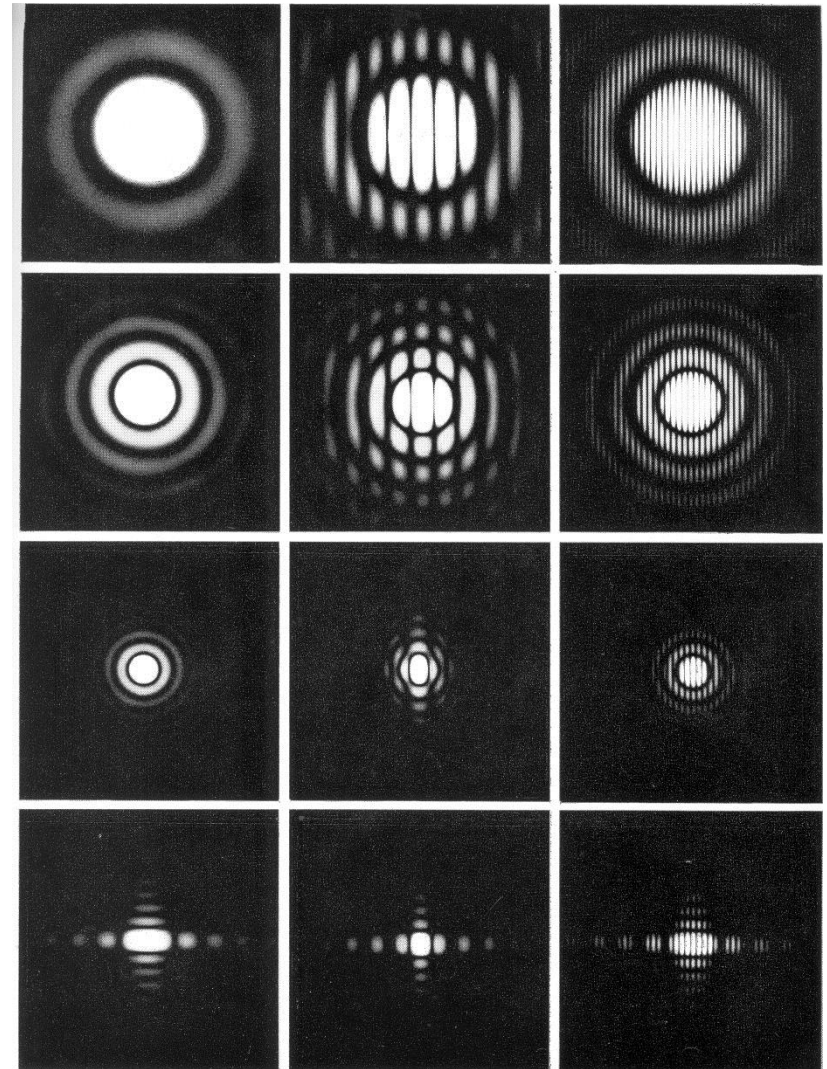
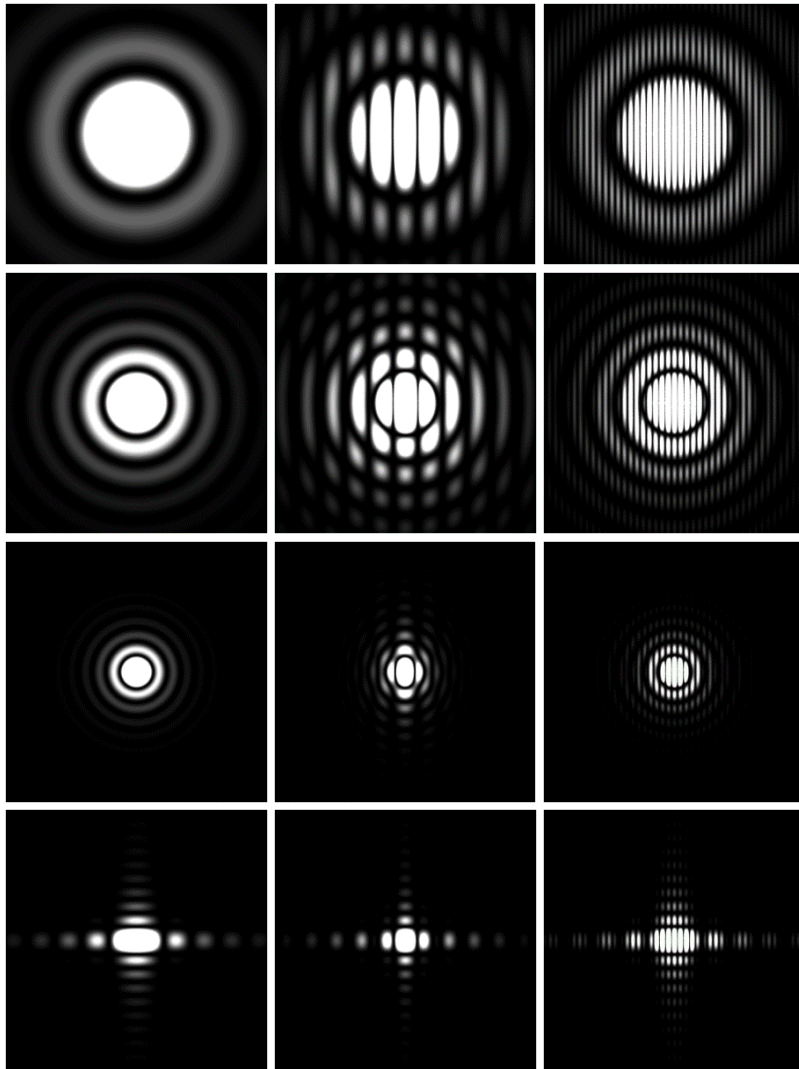


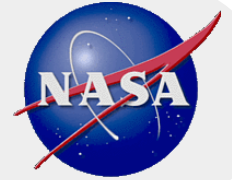
(b) Plate 2





Theory versus Experiment: Plate 1





Theory versus Experiment: Plate 2

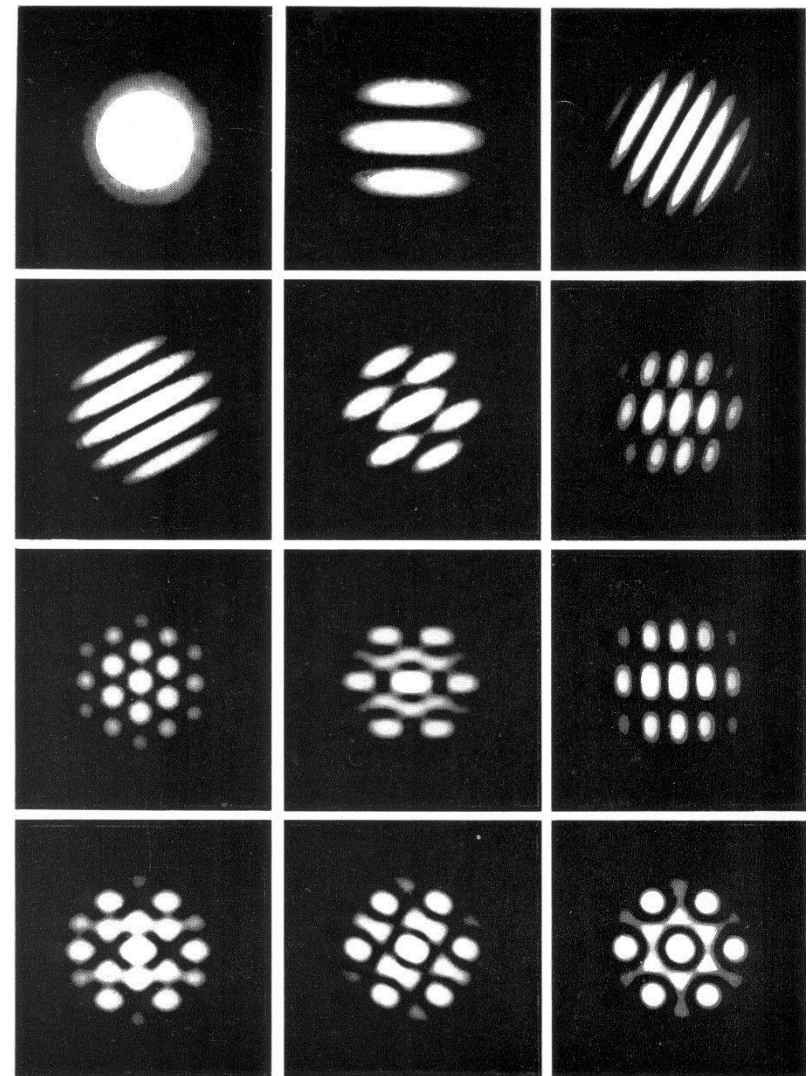
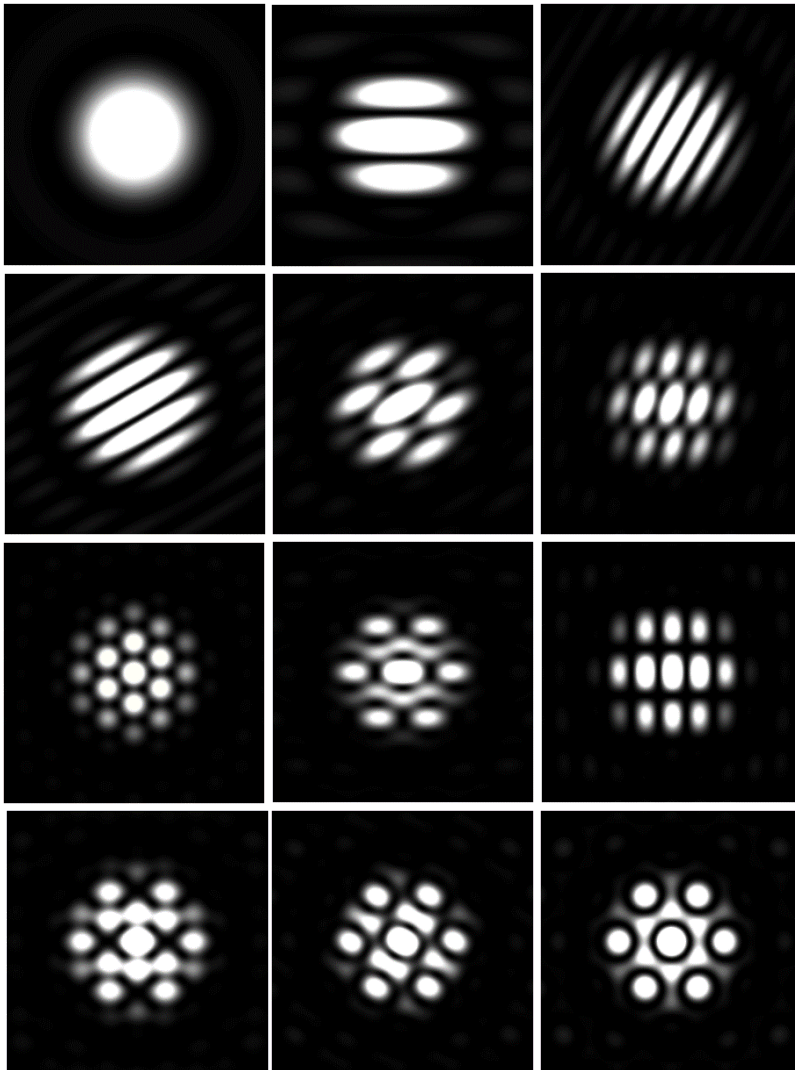
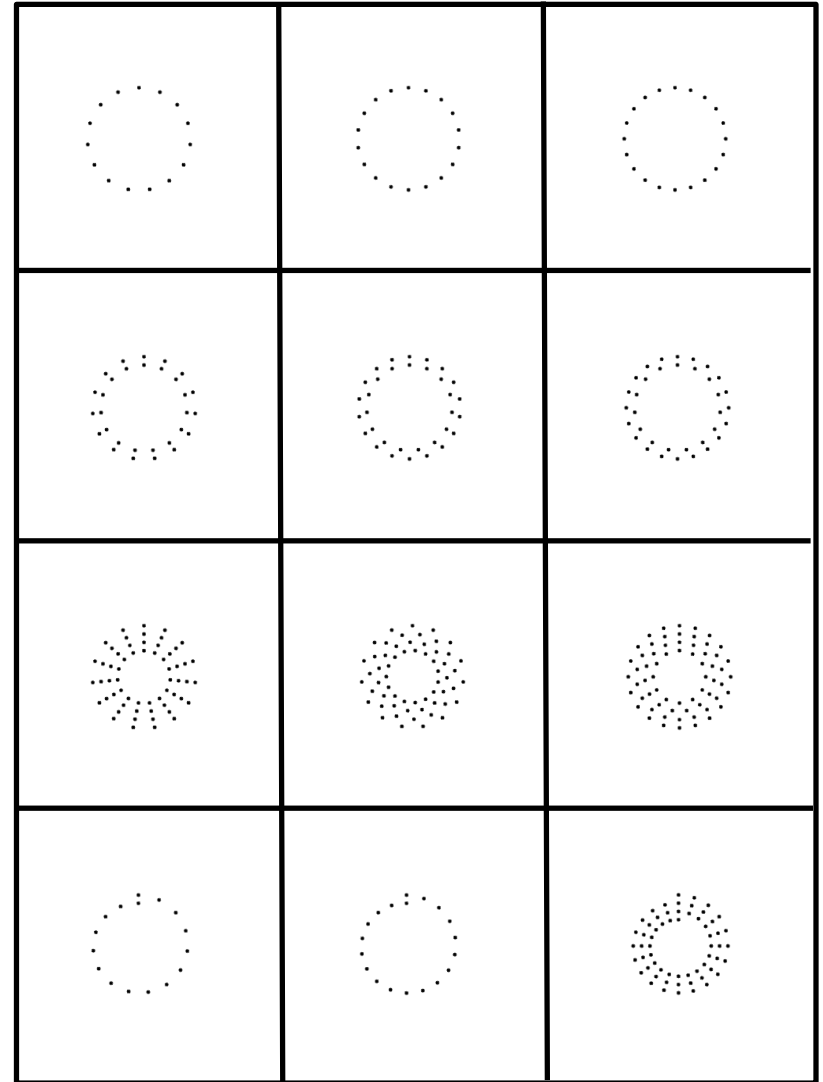
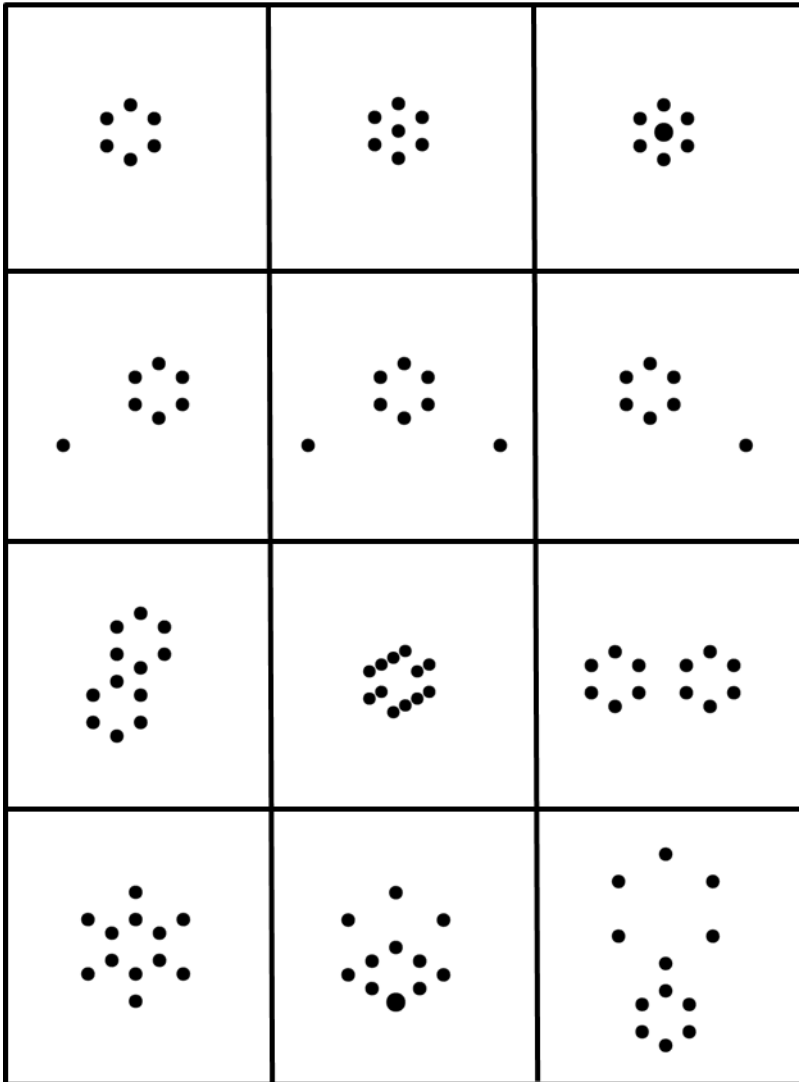
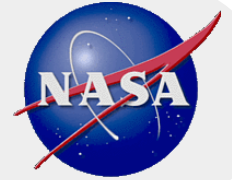


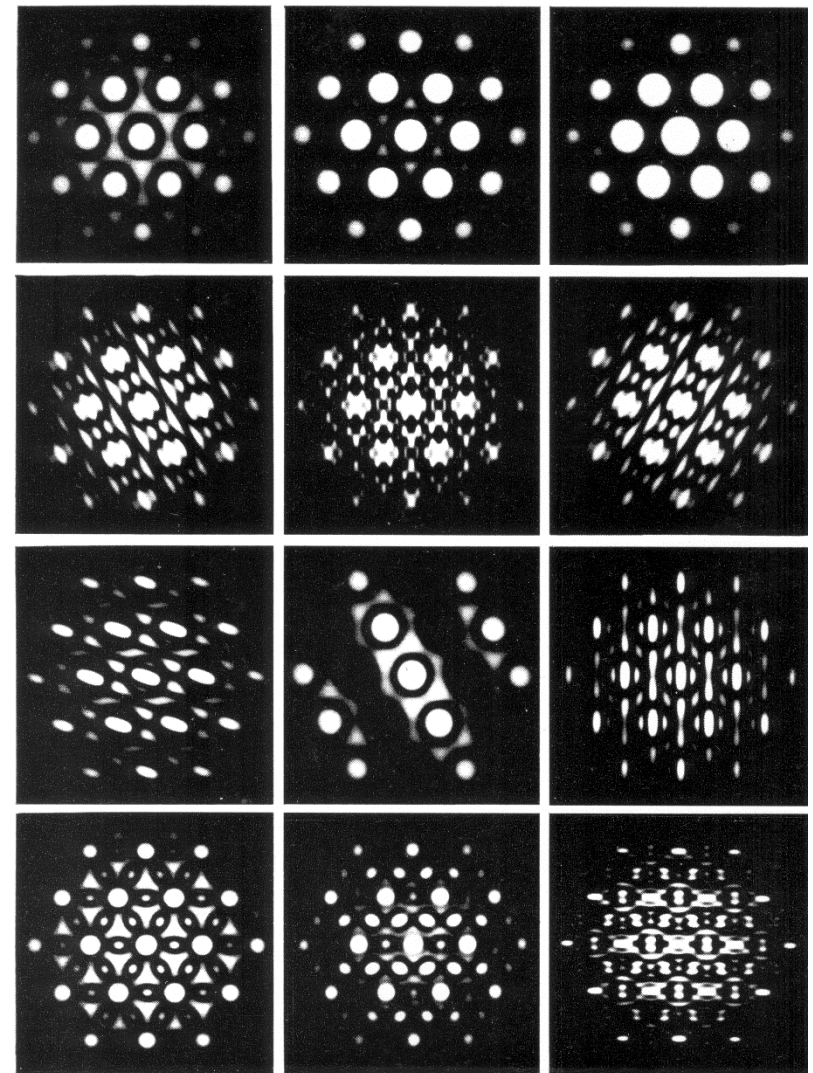
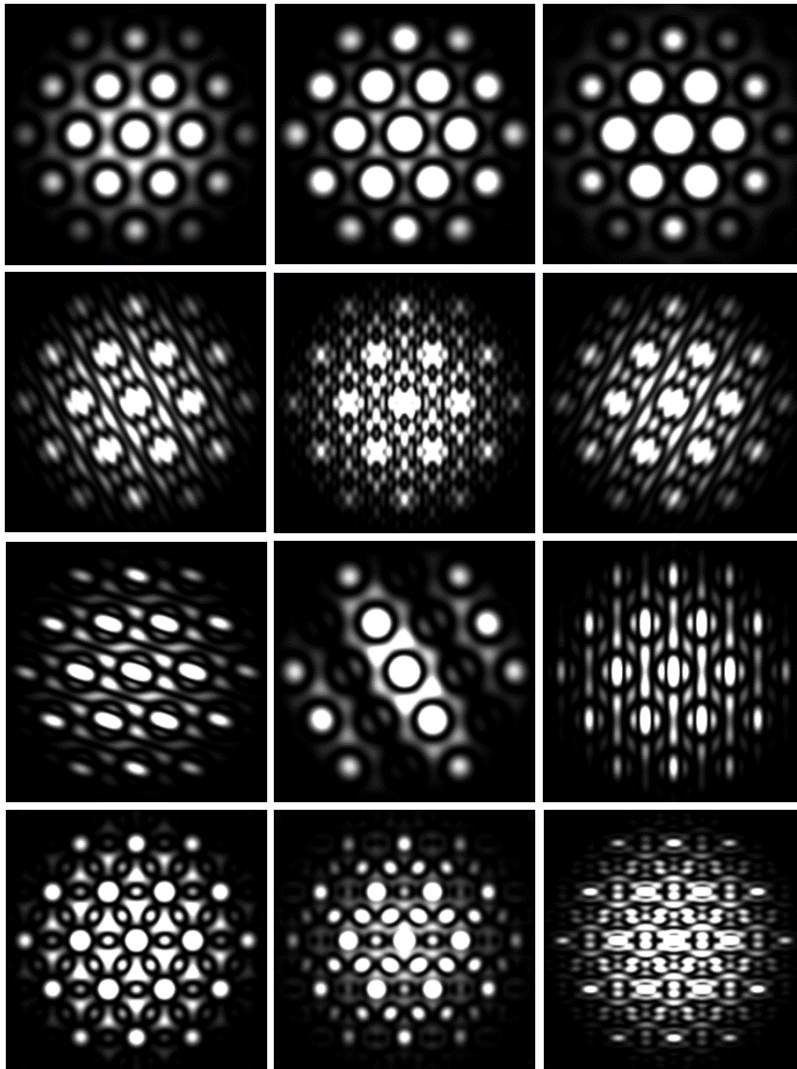


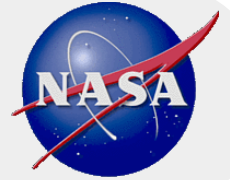
Plate 5 & 14



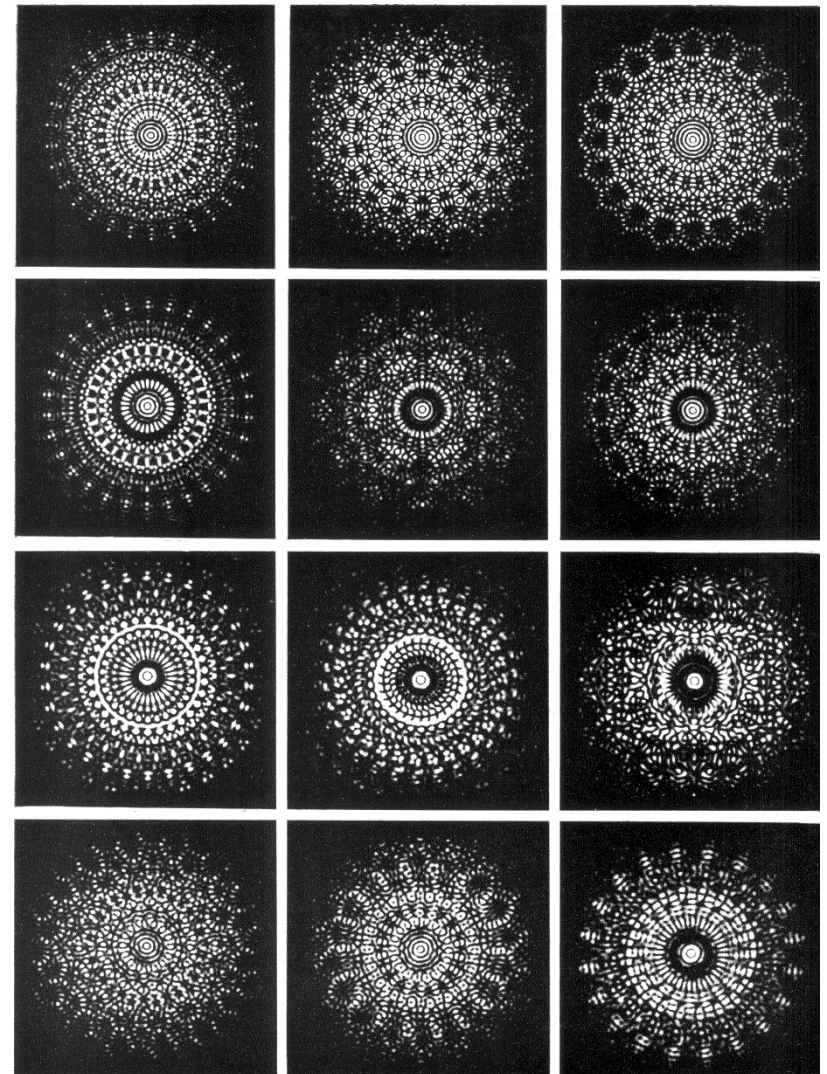
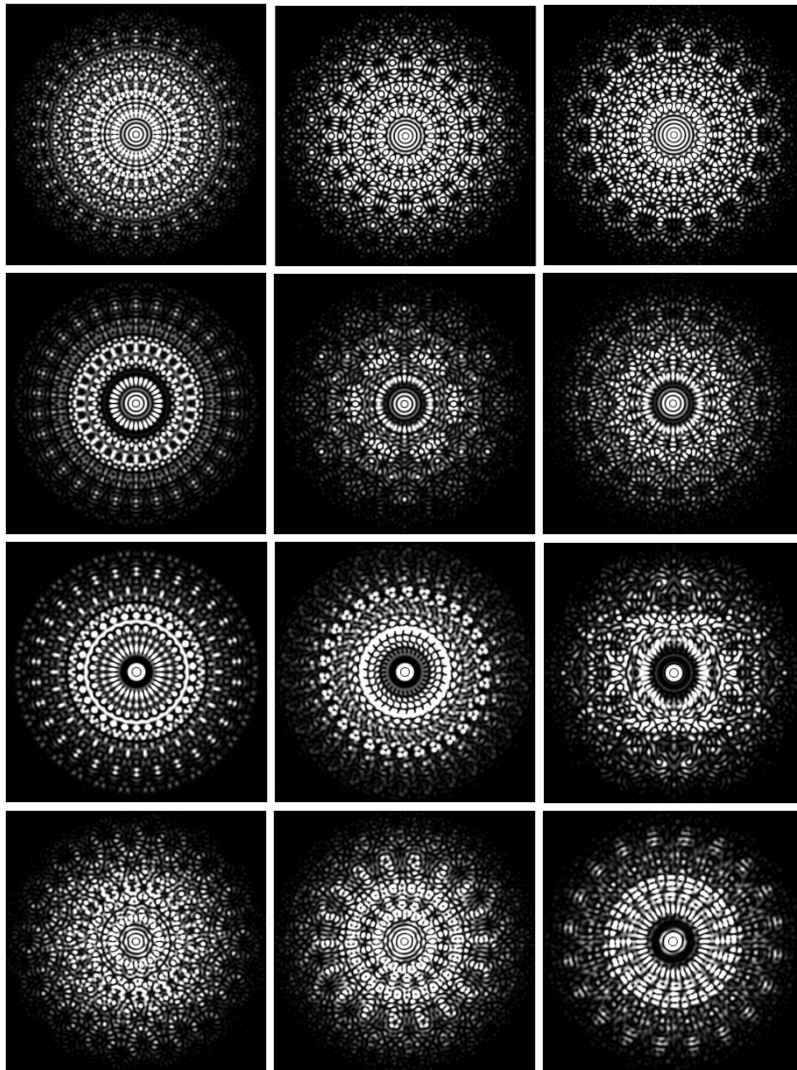


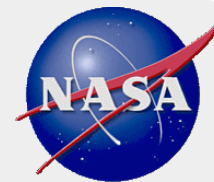
Theory versus Experiment: Plate 5





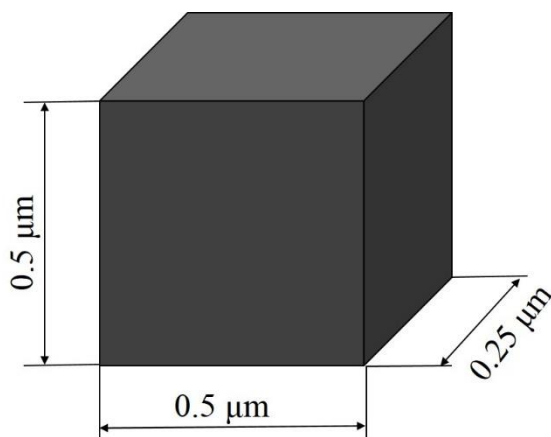
Theory versus Experiment: Plate 14





Comparison of the Predicted Cross Sections from DDA and GMM-PA: A Simple Example

A block of Au



Incident wavelength:

0.5 μm

Bulk refractive index:

(0.9656, 1.863)

DDA replaces the Au block by an array of 131,072 (64 x 64 x 32) dipole scatterers, while GMM-PA uses 131,072 “atomic spheres” of ~ 7.8 nm diameter

| | Q_{ext} | Q_{abs} | Q_{sca} | $\langle \cos\theta \rangle$ |
|--------|------------------|------------------|------------------|------------------------------|
| DDA | 3.625 | 1.451 | 2.174 | 0.422 |
| GMM-PA | 3.627 | 1.456 | 2.171 | 0.414 |

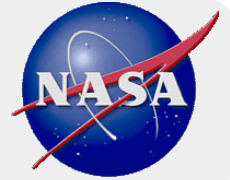
Q_{ext} – Extinction efficiency

Q_{abs} – Absorption efficiency

Q_{sca} – Scattering efficiency

$\langle \cos\theta \rangle$ – Asymmetry parameter

The efficiencies shown are cross sections normalized by the geometrical cross section of the volume-equivalent sphere



Remarks and Discussions (1/2)

- The framework of the GMM-PA scattering formulation has been nearly complete, which is the special version of the Generalized Multi-particle Mie-theory (GMM) for finite periodic arrays (PAs)
 - **Implies an approximation with regard to “edge effect”**
 - **Can handle PAs having enormous (e.g., $\gg 10^6$) component units**
 - **In the atomic theory, matter is regarded as composed of interacting particles (atoms and molecules) embedded in the vacuum**
- GMM-PA solutions to the spatial distribution of a scattered field have been compared with experimentally obtained Fraunhofer diffraction patterns
- Systematic comparisons of the GMM-PA with other scattering theories are underway for the prediction of cross sections (such as for extinction, scattering, absorption, and radiation pressure)



Remarks and Discussions (2/2)

- In the GMM-PA, there are different ways to calculate cross sections, including a highly efficient integral approach to calculating the total scattering cross section and asymmetry parameter
 - **Numerical solutions obtained from different approaches must be consistent**
 - **In the current implementations, a material body is represented by a PA embedded in the matrix of vacuum; practical test calculations seem to suggest that it may be better to include attenuation of electromagnetic waves in the matrix for absorbing materials, especially for electric conductors**
- To solve the multiple scattering from a PA, the complex refractive index of the component scatterers must be provided. When using a PA to characterize a material body, this demands the use of an effective medium theory (EMT), to obtain the required input parameters from those given for corresponding bulk materials
 - **There is no commonly accepted general EMT available at this point**